

Gapless spin-liquid phase in an extended spin 1/2 triangular Heisenberg model

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We numerically study Heisenberg models on modified triangular lattices, and show that a weak next-nearest-neighbor exchange interaction added to a simple triangular lattice, namely, the equilateral triangular lattice with only the nearest-neighbor exchange is sufficient to stabilize a quantum spin liquid against the antiferromagnetic order widely accepted as the ground state of the simple triangular model. The spin gap (triplet excitation gap) and spin correlation at long distances decay algebraically with increasing system size at the critical point between the antiferromagnetic and spin-liquid phases as well as inside the spin-liquid phase, indicating the presence of an unconventional critical (algebraic spin-liquid) phase characterized by the dynamical and anomalous critical exponents $z + \eta \sim 1$. Unusually small triplet and singlet excitation energies found in extended points of the Brillouin zone impose constraints on this algebraic spin liquid.

KEYWORDS: triangular lattice, next nearest neighbor interaction, J_1 - J_2 Heisenberg model, many-variable variational Monte Carlo method, gapless quantum spin liquid, spin gap, spinon Fermi surface

The spin liquid was first proposed by Anderson as a resonating valence bond (RVB) ground state of the spin 1/2 antiferromagnetic Heisenberg model on the triangular lattice.¹⁾ Although it is now widely believed that the true ground state of the model has a long-ranged magnetic order,²⁻⁴⁾ two-dimensional systems based on some modification of the triangular Heisenberg model provide us with realistic possibilities of reaching the spin liquid. This is because some of organic conductors are basically described by the triangular structure with some additional elements and they indeed do not show clear indications of the symmetry breaking.^{5,6)} If one can understand how and in which direction the spin-liquid state is more stabilized and what kind of spin liquids is expected even on the level of model studies, it greatly helps as a clue for stabilizing them.

In this letter, we study the Heisenberg model on the triangular lattice shown in Fig. 1(a) with weak next-nearest neighbor interaction J_2 illustrated in the inset of Fig. 1(b). The Hamiltonian has the form

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where \mathbf{S}_i , $\langle i,j \rangle$, and $\langle\langle i,j \rangle\rangle$ denote the quantum $S = 1/2$ spin at the site i , nearest-neighbor sites, and next-nearest-neighbor sites, respectively.

Previous studies⁷⁻¹⁶⁾ have suggested that the model contains rich phases. For the classical Heisenberg model, the 120° Néel state illustrated in Fig. 1(a) is stable for $J_2/J_1 < 1/8$, while for $J_2/J_1 > 1/8$, the ground states become degenerate for any four-sublattice states that satisfy $\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4 = 0$, where \mathbf{S}_i denotes the spin at sublattice site i . When $J_2/J_1 > 1$, an in-

commensurate spin structure appears. By considering quantum fluctuations perturbatively, stripe-type antiferromagnetic state illustrated in Fig. 1(b)⁸⁻¹⁴⁾ was proposed for $1/8 < J_2/J_1 < 1$. Then the spin-liquid state could emerge around $J_2/J_1 = 1/8$, sandwiched by the 120° Néel and stripe states.^{8,10)} Very recently, by using a variational Monte Carlo (VMC) method, Mishmash *et al.* claimed that the spin-liquid phase with the nodal d -wave symmetry is realized for $0.05 \lesssim J_2/J_1 \lesssim 0.17$.¹⁶⁾ However, they overestimated the spin-liquid phase, since their Huse-Elser type wave functions for antiferromagnetic states are relatively inaccurate. Besides, their wave function favors d -wave type spin liquid due to a limited number of variational parameters.

To reveal the nature of the spin liquid ground states possibly realized in this model, we calculate the ground states and low-energy excitations by using a many-variable variational Monte Carlo (mVMC) method.¹⁷⁾ (See the Supplemental Materials¹⁸⁾ for more details.) We find a spin-liquid phase characterized by the gapless excitations and power-law decay of the spin correlation with the critical and the dynamical exponents η and z , respectively satisfying $z + \eta = 1$ in the parameter region $0.10(1) \leq J_2/J_1 \leq 0.135(5)$, as shown in Fig. 1(b).

Three locally stable states are found as candidates of the ground state; the 120° Néel state, the stripe state, and the spin-liquid state. As shown in Fig. 2, these states are characterized by the spin structure factor defined as

$$S(\mathbf{q}) = \frac{1}{3N_s} \sum_{i,j} e^{i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \quad (2)$$

where \mathbf{R}_i denotes the position vector of the site i . The existence of the long-ranged order is assured when the maximum of $S(\mathbf{q})$ at $\mathbf{q} = \mathbf{Q}$ scales as $S(\mathbf{Q}) \propto N_s$ for large N_s . The ground state is determined from the com-

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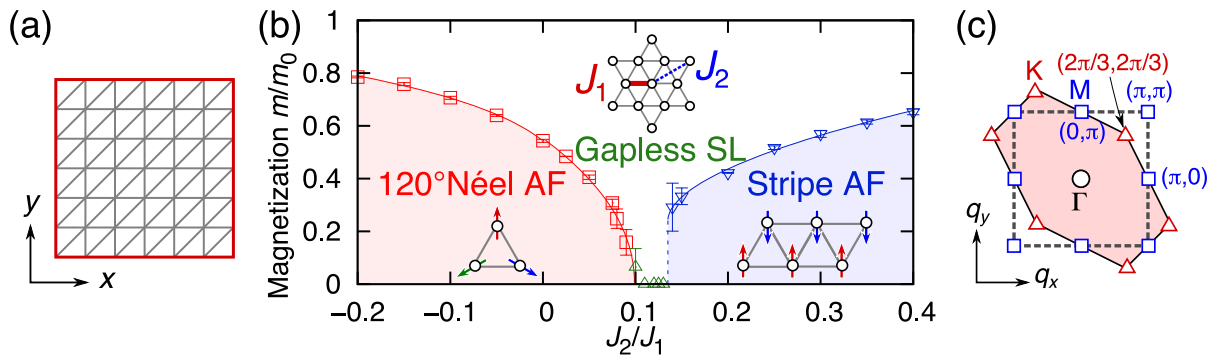


Fig. 1. (color online) (a) Deformed triangular lattice, which is topologically equivalent to isotropic triangular lattice. (b) Phase diagram of antiferromagnetic J_1 - J_2 Heisenberg model on triangular lattice obtained in the present study. The red square, green up-pointing triangular, and blue down-pointing triangular points are order parameters (sublattice magnetizations) of the 120° Néel states (120° Néel AF), spin liquid (SL), and stripe states (Stripe AF), respectively. The lines are guide for the eyes. The magnetization is estimated as $m/m_0 = 2\sqrt{c} \times \lim_{N_s \rightarrow \infty} S(\mathbf{Q})/N_s$, where c denotes a correction factor, and $S(\mathbf{Q})$ is the peak value of the spin structure factor. For the 120° Néel states and the spin-liquid states, $c = 6$, while for the stripe states, $c = 3$. Here, $m_0 (= 1/2)$ is the saturated magnetization expected in the classical Néel order. (c) Brillouin zone of deformed triangular lattice. Equivalent momenta are represented by the same symbols.

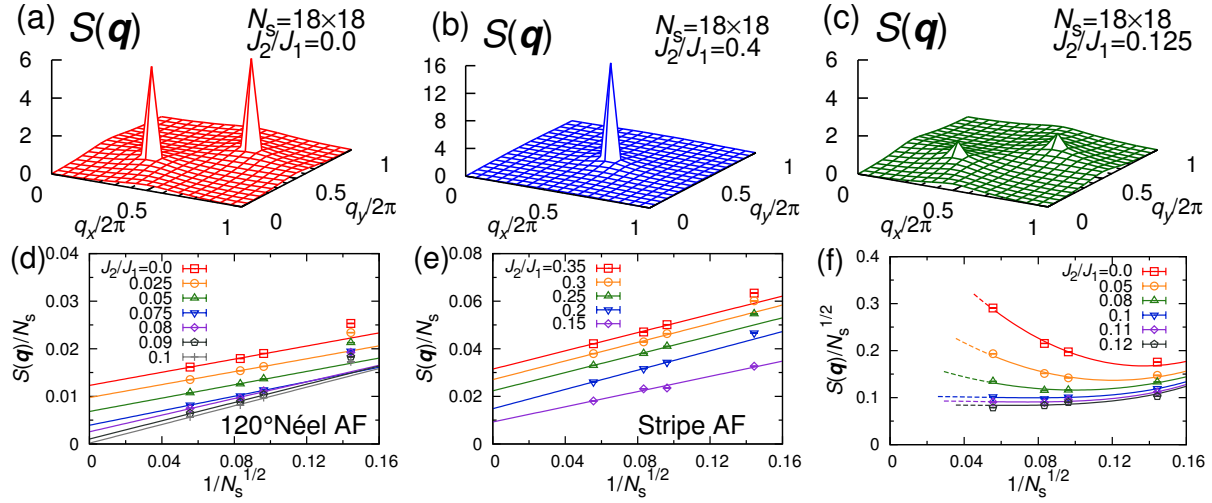


Fig. 2. (color online) (a-c) Spin structure factors ($N_s = 18 \times 18$) for 120° Néel state at $J_2/J_1 = 0$ (a), stripe state at $J_2/J_1 = 0.4$ (b), and spin-liquid state with short-ranged magnetic correlation with the 120° spin alignment at $J_2/J_1 = 0.125$ (c). (d-f) Size dependence of peak value of $S(\mathbf{Q})/N_s$ for 120° Néel states (except $J_2/J_1 = 0.1$) (d), stripe states (e), and $S(\mathbf{Q})/N_s^{1/2}$ for spin liquid (f). Size dependence at $J_2/J_1 = 0.1$ is also shown in (d) for reference. We extrapolate the data by using the relation $S(\mathbf{Q})/N_s = m^2/c + S_1/N_s^{1/2} + S_2/N_s$, where m denotes the extrapolated magnetization, c denotes a correction factor which is the same as Fig. 1(a), while S_1 and S_2 are constants. For antiferromagnetic states, we use the data for $N_s > 100$ to reduce the finite-size effect, and fit them by the relation with $S_2 = 0$ to estimate the statistical error bars. On the other hand, to precisely determine the phase boundary for spin-liquid states, we also examine the size dependence of $S(\mathbf{Q})$. We find that $S(\mathbf{Q})$ scales as $S(\mathbf{Q}) \propto N_s^{1/2}$ for $J_2/J_1 \geq 0.1$.

parison of the energy, which is extrapolated to the thermodynamic limit, if more than one locally stable states are found.

For $J_2/J_1 < 0.1$, we find the 120° Néel state as the ground state. In the Brillouin zone shown in Fig. 1(c), Bragg peaks appear at $\mathbf{q} = \pm(2\pi/3, 2\pi/3)$ corresponding to the 120° spin structure, as shown in Fig. 2(a). From the size extrapolation of $S(\mathbf{Q})/N_s$ in Fig. 2(d), we obtain the magnetization of the 120° Néel state at $J_2/J_1 = 0$ as $m/m_0 = 0.543(6)$, where $m_0 = 1/2$ is the saturated magnetization in spin 1/2 systems. This value is nearly the same as $m/m_0 = 0.53$, which is the best variational result in the literature by Heidarian *et al.*¹⁹⁾ For $J_2/J_1 > 0.135$, we find the stripe ordered state as the ground state. As

shown in Fig. 2(b), it shows a Bragg peak at $\mathbf{q} = (\pi, \pi)$ corresponding to the ferromagnetic spin alignment along the $x = y$ direction and the antiferromagnetic spin alignment along the x and y axes on the lattice shown in Fig. 1(a). The state has long-ranged order, as shown in Fig. 2(e).

Sandwiched by the 120° Néel and stripe states, the spin liquid is stabilized. It shows diffusive peaks at $\mathbf{q} = \pm(2\pi/3, 2\pi/3)$, which are reminiscent of the 120° spin alignment, as shown in Fig. 2(c). We find in Fig. 2(f) that the peak scales as $S(\mathbf{Q}) \propto N_s^{1/2}$ in all the regions of the spin-liquid phase ($0.10 < J_2/J_1 < 0.135$) implying the emergence of a critical phase without the long-ranged order. In fact, this scaling leads to the spin correlation

$$\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \propto 1/|\mathbf{R}_i - \mathbf{R}_j|^{20}.$$

At the transition point $J_2/J_1 = 0.10(1)$, the spin liquid undergoes a continuous quantum phase transition to the 120° Néel state, while at $J_2/J_1 = 0.135(5)$, a level cross to the stripe state occurs.

To get further insight into the nature of the spin-liquid ground state, we study the energies of the momentum-resolved lowest triplet and singlet excited states. We take the total momenta along the symmetric Γ -K-M line, namely $\mathbf{K} = (q, q)$ with $0 \leq q \leq \pi$, as illustrated in Fig. 1(c). The ground states have turned out to be always the singlet and stay at the Γ point irrespective of J_2 and N_s .

According to the spin wave (SW) analyses,^{21,22)} the spin gap Δ for antiferromagnetic states should scale as $\Delta(N_s) \propto 1/N_s$. However, when both magnetic order and spin gap become zero, especially on continuous transition points, more subtle scaling dominates. For example, for the two-dimensional quantum Heisenberg model, when a quantum phase transition occurs between the phase with $O(3)$ long-ranged order and the disordered phase with a finite-gap spin excitation, the nature of the transition was proposed to be essentially the same as that of the two-dimensional quantum $O(3)$ nonlinear sigma model.²³⁾ Since the correlation length is the same in the spatial and time directions, the dynamical exponent z becomes $z = 1$ at the critical point. Therefore, the spin gap, which scales as $\Delta \sim k^z$ at the critical point,²⁴⁾ obeys the relation $\Delta \sim k \propto 1/L$ with $L \equiv N_s^{1/2}$. Later on, a possible deconfined criticality was proposed in that case.^{25,26)}

In general, the critical state may be characterized by the power-law decay $\Delta \propto 1/L^z$ with the dynamical exponent z . As shown in Fig. 3, the spin gap at the critical point ($J_2/J_1 = 0.1$) consistently shows power-law decay for larger sizes although it is difficult to estimate the critical exponent of the decay z quantitatively. In finite size systems, higher order corrections in the extrapolation of the spin gap are non-negligible, that makes the extrapolation difficult. Indeed, in the previous study, the spin gap of the spin liquid on the triangular lattice system shows size dependence as a concave function of $1/N_s$ if the system size is not sufficiently large,²⁷⁾ although the chiral perturbation theory for the square lattice with the well-established antiferromagnetic order predicts the scaling by $1/N_s$ with the negative coefficient of the $1/N_s^{3/2}$ correction.²¹⁾ Therefore, the estimated critical exponents may become larger than the exact exponents.

This critical point turns out to be unusual, because the spin gap appears to decay as power law even deeply inside the spin-liquid phase. Within the available system size ($\leq 18 \times 18$), these results support that the whole spin-liquid phase may belong to the critical phase. This is consistent with the above critical behavior of the spin correlation, because we expect $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \propto 1/|\mathbf{R}_i - \mathbf{R}_j|^{d+z+\eta-2}$ with $d = 2$ and $z + \eta = 1$ in the whole spin-liquid phase as we detail later.

We now study the momentum resolved spectra of the excitations $\Delta(\mathbf{K})$. Generally, if the ground state has a magnetic long-ranged order, corresponding to the

magnon modes, it shows gapless excitations at the Γ point as well as at the wave vectors \mathbf{Q} of $S(\mathbf{Q})$ peaks. We have indeed confirmed that the present mVMC calculation reproduces this expectation.

In the spin-liquid phase, we examine the size dependence of singlet and triplet gaps at $\mathbf{K} = (n\pi/3, n\pi/3)$ ($n \in \mathbb{Z}$) as shown in Fig. 4(a), which can be calculated for all system sizes we studied. As shown in Fig. 4(b) and (c), we find that both singlet and triplet gaps at all the available momenta, systematically and substantially decrease with increasing system size and natural extrapolations suggest vanishingly and unusually small values if it is nonzero. Even at $\mathbf{K} = (\pi/3, \pi/3)$, where the gap is the largest, a simple linear extrapolation suggests the gap $< 0.2J_1$. The gaps at other momenta are likely to be less than $0.1J_1$. To our knowledge, such small values of gap in the extended points of the Brillouin zone are not known. In the antiferromagnetic phase, the spin wave excitation usually has dispersions of the order of the exchange coupling J_1 .

Now, we discuss comparisons with previously proposed spin-liquid states. In the present result, the singlet and triplet excitation energies look vanishingly small at all the studied total momenta including the Γ , K, M, and the middle of the Γ and K points, as shown in Fig. 4(a). If the excitation is gapless at the momenta in a finite area of the Brillouin zone, it can be accounted for by the presence of a large spinon Fermi surface.^{28,29)} Although the present model does not contain itinerant electrons, if the finite-size spin gap scales as $1/L$, the present spin-liquid state is indeed similar to the expected gap scaling of the lowest particle-hole (Stoner) excitation in the (spinon) Fermi liquid. However, the exponent of the power-law decay of the spin correlation ($\sim 1/r^\alpha$ with $\alpha = 1$) contradicts the expected behavior in the presence of the Fermi surface, namely $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \propto 1/|\mathbf{R}_i - \mathbf{R}_j|^3$.³⁰⁾

When the gap closes only at $\mathbf{K} = (n\pi/3, n\pi/3)$ ($n \in \mathbb{Z}$) or one of them it is consistent with the algebraic spin liquid. For example, for the Dirac spin liquid whose dispersion scales as $\Delta \propto k^z$ with $z = 1$, it was discussed that the spin correlation may decay as $1/r^{1+\eta}$ typically with nonzero η because of coupling of Dirac spinons to a noncompact $U(1)$ gauge field. (See^{31,32)} and references therein.) The couplings depend on the flux patterns of the slave-boson mean-field states and hence the exponent η varies according to lattice models; for instance, it was proposed that $\eta \sim 0.5$ for the projected d -wave BCS state³²⁾ and $\eta \sim 3$ for the $U(1)$ Dirac spin liquid on the kagome lattice.³³⁾ More generally, it can be conjectured that the algebraic spin liquid is characterized by the $1/L^z$ scaling of the gap closing and the $1/r^{z+\eta}$ decay of the spin correlation. If $z + \eta = 1$, it is consistent with the present result. The vanishingly small excitation energies at extended momentum points naturally lead to the power-law decay of the spin correlation in real space at the period of the corresponding momentum points. However, so far, the clear algebraic $(1/r)$ decay of the spin correlation is found only for that at the period $(2\pi/3, 2\pi/3)$ (120 degree order point). This rules out the possibility of d -wave type spin liquid,^{16,34)} whose gap is open at the period $(2\pi/3, 2\pi/3)$. In the

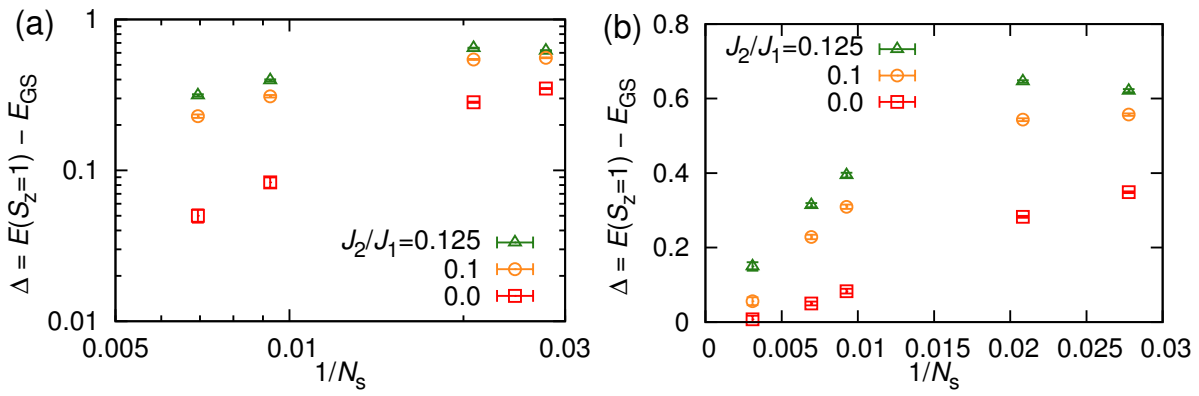


Fig. 3. (color online) Size scalings of spin gaps of 120° Néel state, spin liquid, and near the critical point. The spin gaps at the Γ point are plotted on a log-log plot (a) and a linear plot (b).

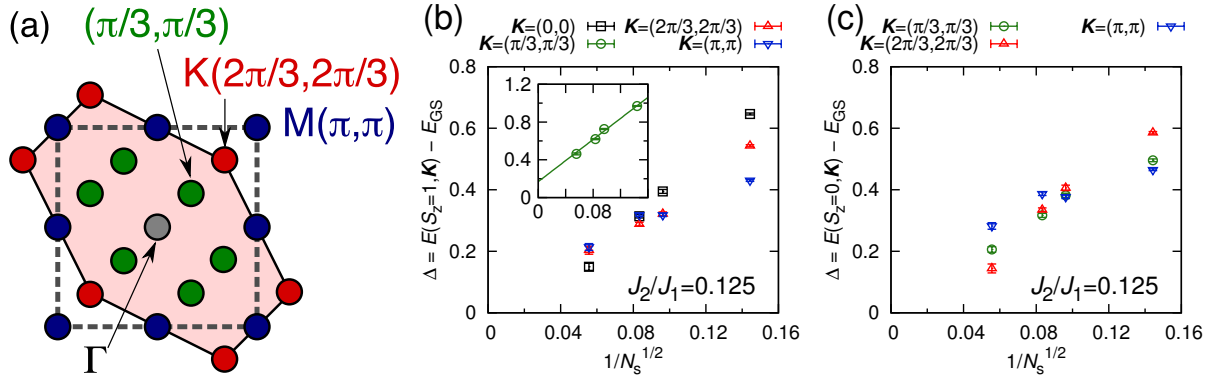


Fig. 4. (color online) Momentum-resolved excitation spectra in spin-liquid phase. (a) Momenta studied for excitation spectra in colors same as (b) and (c). Size dependences of triplet (b) and singlet (c) gaps of each total momenta for spin-liquid phase ($J_2/J_1 = 0.125$). Triplet excitations are gapless only at the Γ and K points for the 120° Néel state while they are gapless only at the Γ and M points for the stripe state. For the spin liquid, in addition to the K , M , and Γ points, the middle of Γ and K points show unusually small excitation energies.

present calculation, the spin correlations of the period at other momentum points decay at least faster than $1/r^2$ because $S(\mathbf{q})$ is at most logarithmically divergent and is likely to stay finite in the thermodynamic limit. If this picture applies so that some of the spin correlations at the periods other than $(2\pi/3, 2\pi/3)$ decay exponentially, we expect the opening of gaps at the wavenumber away from the crossing points. So far the examination of this prediction is not numerically easy in the present computer power. This is left for further studies. Within the available results, the present spin liquid does not contradict the possible algebraic spin liquid or flux state, while our results impose constraints on its nature.

We have shown that the spin-1/2 Heisenberg model on the triangular lattice with the nearest and next-nearest exchange interactions shows an unusual spin liquid, where the spin correlation decays as $1/r$ and the finite-size gap closes as power law as if the quantum critical point is extended to a critical phase. Unusually small gaps at all the total momenta studied here, however, impose a severe constraint on the nature of the spin liquid found in the present study. Clearly, further studies are needed for more unified understanding of the spin-liquid states found in the present study.

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The mVMC codes used for the present computation are based on that first developed by Daisuke Tahara, and that extended by S. M. and Moyuru Kurita. To compute the Pfaffian of skew-symmetric matrices, we employ the PFAPACK.³⁵⁾ The authors thank Takahiro Misawa for fruitful discussions. This work is financially supported by MEXT HPCI Strategic Programs for Innovative Research (SPIRE) and Computational Materials Science Initiative (CMSI). Numerical calculation was partly carried out at the Supercomputer Center, Institute for Solid State Physics, Univ. of Tokyo. Numerical calculation was also partly carried out at K computer at RIKEN Advanced Institute for Computational Science (AICS) under grant number hp120043, hp120283 and hp130007. This work was also supported by Grant-in-Aid for Scientific Research (No. 22104010, and No. 22340090) from MEXT, Japan.

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Supplemental Materials to “Gapless spin-liquid phase in an extended spin 1/2 triangular Heisenberg model”

Variational Monte Carlo method

We consider the variational wave function in the form $|\Psi\rangle = \mathcal{P}\mathcal{L}|\Psi_0\rangle$ with

$$|\Psi_0\rangle \equiv \left(\sum_{i,j=1}^{N_s} \sum_{s,t=\uparrow \text{ or } \downarrow} f_{ij}^{st} c_{is}^\dagger c_{jt}^\dagger \right)^{N_e/2} |0\rangle, \quad (1)$$

which can describe antiferromagnetic states with collinear, noncollinear, and even noncoplanar spin alignment, and also gapless and gapped spin-liquid states on an equal footing. Here, $|\Psi_0\rangle$ is a generalized Bardeen-Cooper-Schrieffer (BCS) wave function, where f_{ij}^{st} and $|0\rangle$ denote the variational parameter and the vacuum. This is a natural extension of the Liang-Doucot-Anderson type wave function¹⁾ and the Hartree-Fock-Bogoliubov type wave function.^{2,3)} Note that not only the product of the paired singlet but also the paired triplet component are allowed in this scheme beyond the RVB singlet,¹⁾ which is crucial in representing non-collinear spin order. We assume f_{ij}^{st} to be long-ranged and allow symmetry breaking, namely six-sublattice structure. To restore the translational symmetry of $|\Psi_0\rangle$, we consider the total momentum projection

$$\mathcal{L}^{\mathbf{K}} = \frac{1}{N_s} \sum_{\mathbf{R}} e^{i\mathbf{K}\cdot\mathbf{R}} T_{\mathbf{R}}, \quad (2)$$

where \mathbf{K} and $T_{\mathbf{R}}$ denote the total momentum [defined in Fig. 1(c)] and the translation operator, respectively. We apply the projection onto all the possible total momenta. In addition to the Gutzwiller factor

$$\mathcal{P}_G = \prod_i (1 - n_{i\uparrow} n_{i\downarrow}) \quad (3)$$

with $n_{is} = c_{is}^\dagger c_{is}$, which excludes the double occupation of the electron, to reach a wave function closer to the true ground state (lowest triplet state), we also impose a projection $\mathcal{P}_{S_{\text{tot}}^z=0}$ ($\mathcal{P}_{S_{\text{tot}}^z=1}$) on the wave function by keeping the difference in the number of up and down spins fixed as zero (two). We consider lattices for $N_s = L \times L$ and $\sqrt{3}L \times \sqrt{3}L$ under the periodic boundary condition. By using the stochastic reconfiguration (SR) method,⁴⁾ we optimize the parameters by typically 2000 SR steps. After confirming the energy convergence, we calculate the

expectation values of the physical quantities and average them over more than 10 independent runs containing typically 2000 Monte Carlo steps each to estimate statistical errors.

Spin correlation function in spin-liquid phase

Here, we discuss the spin correlation function in the spin-liquid phase. In the Ginzburg-Landau theory, the spin correlation function at the critical point is given as $C(r) \propto r^{-(d-2)}$, where $d(> 2)$ is the spatial dimension. Beyond the Ginzburg-Landau theory, at the critical point and inside the critical phase, the correction may be given by the dynamical exponent z and the anomalous dimension η as $C(r) \propto r^{-(d+z+\eta-2)}$ ($d+z+\eta > 2$).^{5,6)} This

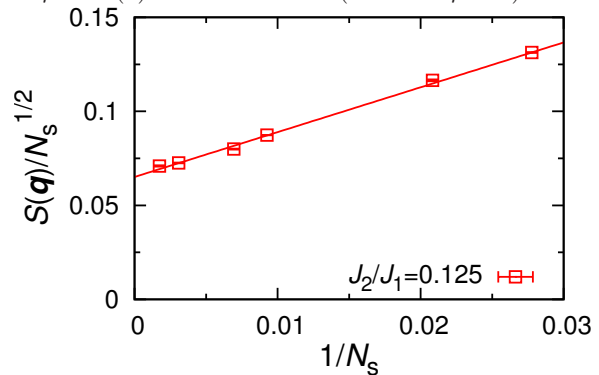


Fig. S1. (color online) Size dependence of peak value of spin structure factor divided by square root of system size. It is well fit by the scaling $S(q)/N_s^{1/2} = S_1 + S_2/N_s$, where S_1 and S_2 are constants.

also leads to $S(Q, N_s) \propto N_s^{2-(d+z+\eta)/2}$ for $d+z+\eta < 4$. Figure S1 shows that the spin liquid follows the critical scaling with $z+\eta \sim 1$.

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